

ULTRASONIC WELDING ON Cu-Zr BASED GLASSY RIBBONS

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Novel joining techniques for ribbons with amorphous structures are developed in order to implement them in new products. Having a chemical composition that is CuZr-based they found use in various engineering fields. In order to diversify the applicability of the ribbons, ultrasonic welding is used. Ribbons having the nominal chemical composition Cu₄₅Zr₄₅Al₅Ag₅ and Cu₄₀Zr₄₅Al₅Ag₁₀ have good mechanical properties and good glass forming ability which make them part of a new class of materials. The arc-melter technique was used to produce the master alloy, consequently ribbons were made using the melt spinning technique and afterwards welded using an ultrasonic equipment. The structure was investigated using differential scanning microscope, X-ray diffractometry. Welded areas were investigated using scanning electron microscope. Thus, ribbons are investigated in order to see if the chemical composition analyzed in this study is a good candidate and a novelty for ultrasonic welded products.

Keywords: glassy ribbons, melt spinning technique, ultrasonic welding

1. Introduction

Glassy state materials raised interest in the engineering field for their properties which are superior to those of crystalline structures. In order to increase the applicability of glassy state materials ultrasonic welding was introduced as a way of joining such materials. In the electronics field this method is used for wire connections with lead frames and pads on semiconductor-microchips or power devices. As years passes by this joining technique became used for electronic packaging because of its flexibility feature for when design changes are needed and the low cost that it implies [1]. Welding products with a glassy structure is a problematic and delicate task. But a large interest in overcoming this issue was

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manifested and various techniques were developed: electron beam welding [2], explosive welding [3], pulse current welding [4], friction welding [5], laser welding [6-9] and ultrasonic welding [10]. Using ultrasonic welding as a way of joining products with a glassy structure arises some difficulties of its own. Among them, preserving the glassy structure in the welded area and the number of parameters that is limited [11]. Weldability of the desired products is described by the dimensions and glass forming ability (GFA) [12]. But there are advantages to laser welding: it can produce bulk metallic glasses (BMG) when overlapped ribbons are welded, thus various shapes or thicknesses of BMG can be obtained. Another convenience of the method is the large spectrum of materials that can be welded by it: dissimilar materials, metallic glasses and nanocrystalline alloys [13]. For this study Cu-Zr based alloys were used. Previous reports show that these alloys exhibit high wear resistance and their mechanical properties are renowned for their high elastic limit, large ductility in bending, high strength [14] and large compressive plastic strain [15]. In order to increase their GFA and improve the mechanical properties Al and Al-Ag were added to the chemical composition [16-17]. The nominal chemical compositions are $\text{Cu}_{45}\text{Zr}_{45}\text{Al}_5\text{Ag}_5$ and $\text{Cu}_{40}\text{Zr}_{45}\text{Al}_5\text{Ag}_{10}$. The study consists in testing and evaluating the possibility of ultrasonic welding when Al and Al-Ag is added to the glassy Cu-Zr based ribbons.

2. Experimental procedures

The master alloys were made using the vacuum arc remelting technique using a MRF ABJ 900 equipment. Chemical elements with a purity of 99.9% were added in the chamber of the installation and using a water-cooled tungsten electrode melted in a button shaped all the elements. The procedure was done in controlled vacuumed atmosphere. In order to ensure a chemical homogeneity and a rigorous mixing of the constituting elements the master alloys were remelted three to four times. Ribbons were produced using the melt spinning technique in an open environment. In order to eject the molten alloy through the quartz crucible (Ilmasil PN) on the rotating copper wheel, argon pressure was used. To avoid the formation of oxides, the area between the tip of the crucible and the copper wheel was protected with an extra source of argon. The results were ribbons with a thickness of $20\mu\text{m}$ and a width of 2 mm. The ultrasonic equipment has a 3000 W generator and resonates at a frequency of 20 KHz. The micro vibrations are concentrated into an ultrasonic horn, made of C45R quality steel, heat treated, with specific geometry to the materials to be joined [13]. In Fig. 1, the ultrasonic welding concept is illustrated. In order to ensure the weld, the amplitude was set on $20\text{ A}/\mu\text{m}$, the welding time was 0.3 T/s, the pressing force was 2.1 mBar and the frequency was 20 f/kHz.

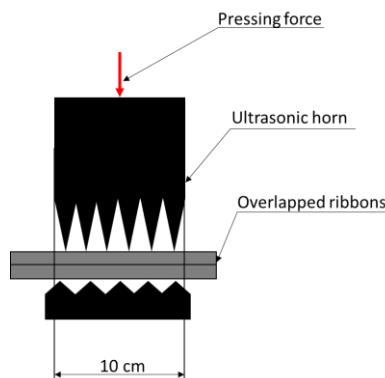


Fig. 1. Ultrasonic welding concept

Several batches of ribbons were welded by overlapping them, from two to five ribbons, one on top of the other. For this study for each chemical composition five ribbons were overlapped. In Fig. 2 the outcome of the ultrasonic weld is represented. On a macroscopic level it can be seen clearly that the ultrasonic weld was a success on the Cu-Zr based alloys. For further studies the ribbons can be welded in different complex shapes or even mix batches of different chemical compositions.



Fig. 2. Macro images of ultrasonic welded ribbons (upper view and cross-section view)

The microstructure examination on welded area was done using FEI Inspect-S scanning electron microscope. Chemical characterization of the ribbons was done using the EDAX Ametek Element module. Structural characterization of the as-cast ribbon was realized using X-ray diffractometry with a Co-K α radiation ($\lambda=1.78901$ nm) operated at 40kV and 40mA on a Philips PW3064. DSC analysis was done on a NETZSCH STA 449F1 equipment using a heating rate of 20 K/min and the temperature interval was from 30 °C to 700°C in an Al₂O₃ crucible under N₂ atmosphere.

3. Results and discussion

In order to produce glassy ribbons, firstly, master alloys were produced using the arc-melter furnace. Two batches of button shaped master alloys were produced for each chemical composition.



Fig. 3. Arc melting furnace MRF ABJ 900



Fig. 4. Master alloys for $\text{Cu}_{45}\text{Zr}_{45}\text{Al}_5\text{Ag}_5$ (a) and $\text{Cu}_{40}\text{Zr}_{45}\text{Al}_5\text{Ag}_{10}$ (b)

Prior to the ultrasonic weld of the ribbons, a structural analysis was done on the as-cast ribbons. In Fig. 5 is represented the diffractogram from X-ray diffractometry. The shape of the broad diffraction maxima can be clearly seen suggesting the glassy structure of the ribbons. No crystalline peaks are observed suggesting that the melt spinning technique in an open environment provides a good alternative of producing glassy ribbons.

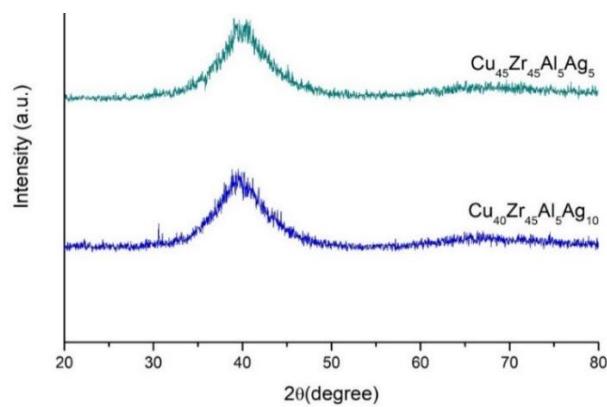


Fig. 5. XRD of as-cast ribbons

The structure of the welded area was investigated. In Fig. 6 it is shown the diffractogram of the welded area. The broad diffraction maxima is retained thus suggesting the glassy state is preserved even when an ultrasonic weld is done. The heat generated from the process, and the pressure from the ultrasonic horn does not affect the structure of ribbons.

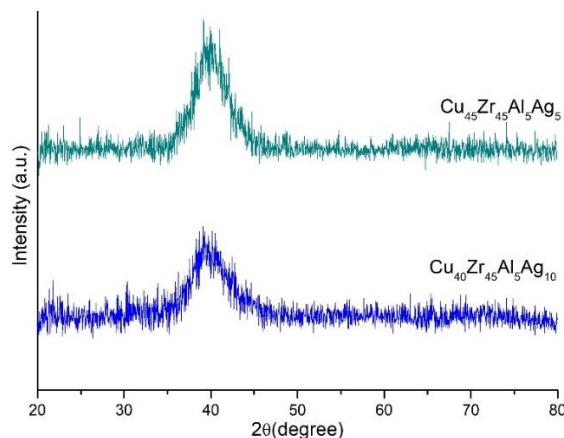


Fig. 6. XRD on welded area

Fig. 7 shows results from the DSC analysis of the glassy ribbons, before the ultrasonic weld. Upon heating it is revealed the following temperatures: glass transition succeeded by crystallization temperature (suggested by the exothermic peak).

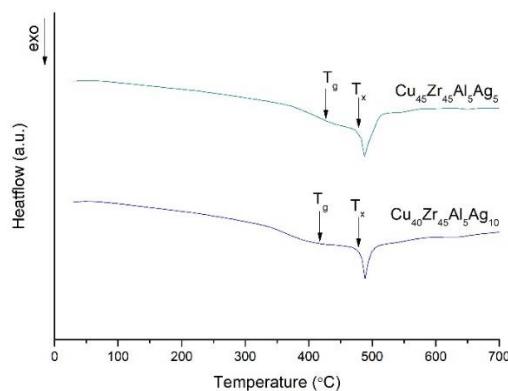


Fig. 7. DSC result for as cast ribbons

After the ultrasonic weld, from each chemical composition, the welded area was subjected to another DSC analysis. In order to do a comparison, the inputs were kept the same. The glass transition temperature and the crystallization

temperature values are inserted in table 1. From Fig. 8 it is noted that no significant structural modifications occurred this is also confirmed by the values.

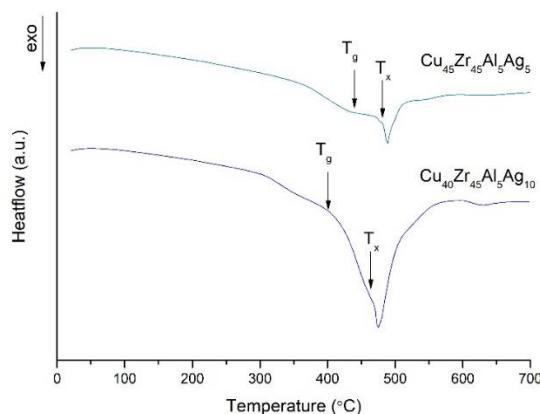


Fig. 8. DSC result after ultrasonic welding

Comparison of DSC results between as-cast and ultrasonic welded ribbons

Table 1

Material	T_g [C]		T_x [C]	
	as cast	ultrasonic weld	as cast	ultrasonic weld
$\text{Cu}_{45}\text{Zr}_{45}\text{Al}_5\text{Ag}_5$	372	372	484	484
$\text{Cu}_{40}\text{Zr}_{45}\text{Al}_5\text{Ag}_{10}$	343	345	485	471

The welded ribbons were hot embedded and afterwards grinded and polished so as to obtain a clear image of the cross section on how to bond formed between the glassy ribbons. Figs. 9 show SEM micrographs of the welded areas on magnifications of 1000x. As it can be seen in Fig. 9, in the case of $\text{Cu}_{45}\text{Zr}_{45}\text{Al}_5\text{Ag}_5$ sample all five ribbons have been welded and the adherent interfaces appear to have almost the same color of the ribbons. The untreated surfaces of the ribbons were enough to ensure a good bonding of the ribbons. For this study, regarding the $\text{Cu}_{40}\text{Zr}_{45}\text{Al}_5\text{Ag}_{10}$ sample (Fig. 9.b) it appears that only four out of five ribbons were successfully welded. This was not the case when two or more ribbons were ultrasonic welded. This is due to either improver fixing in the ultrasonic equipment or maybe one ribbon changed its position and thus its full width was not caught in the process. The process was repeated and on a macroscopic level it appears that all five ribbons were bonded. But in the case of the four ribbons the bond appears to be successful.

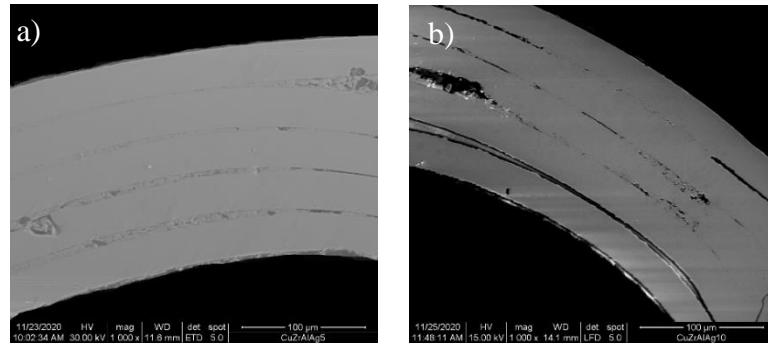


Fig. 9. Cross Section SEM on Cu₄₅Zr₄₅Al₅Ag₅ (a) and Cu₄₀Zr₄₅Al₅Ag₁₀ (b)

The EDX spectrum from Fig. 10 confirms the presence of all chemical elements. Zr having a high reflected characteristic X-ray energy has higher peaks than Cu, Al or Ag.

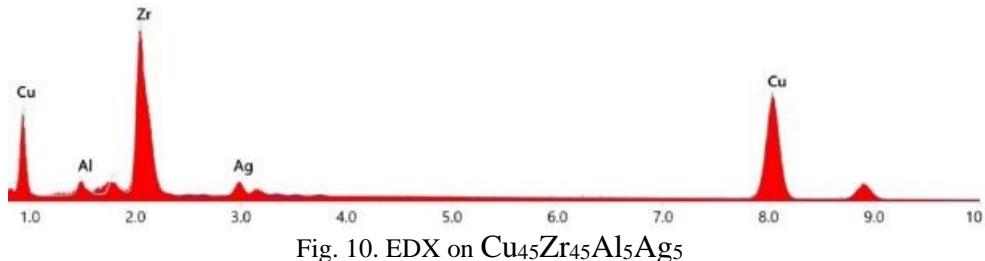


Fig. 10. EDX on Cu₄₅Zr₄₅Al₅Ag₅

Regarding the sample with Ag₁₀ (Fig. 11), EDX also confirms the presence of all chemical elements, but a higher peak is observed for Ag due to higher percentage in the composition.

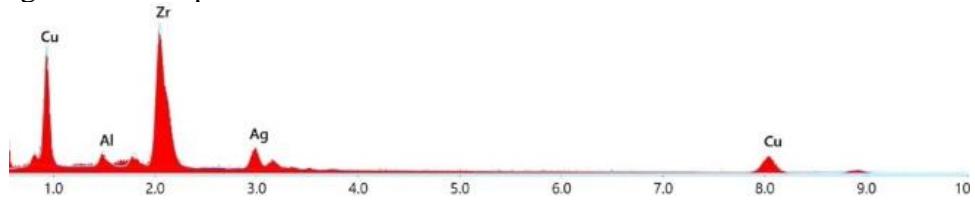


Fig. 11. Cross Section SEM on Cu₄₀Zr₄₅Al₅Ag₁₀

6. Conclusions

After successfully producing the master alloys, the glassy ribbons were made using the melt spinning technique in an open environment. DSC and XRD results confirm their glassy structure. Multiple overlapped ribbons can be welded using the ultrasonic welding equipment. The structure was not affected, as shown in the XRD and DSC results that were realized after the weld. The values of the glass transition and the crystallization temperatures suffered a slight decrease after

the weld in the case of the Cu₄₀Zr₄₅Al₅Ag₁₀ ribbons. The heat generated by the ultrasonic horn, or the pressure applied by it did not affect in any way the structure of the ribbons. SEM micrographs show the existence of metallic bonding between the ribbons. Further studies will consist in welding ribbons in different complex shapes in order to be inserted as reinforcement in composite structures. This will improve the composites mechanical properties as it was reported in previous studies but using different chemical compositions and shapes. Another aspect to study is whether applying external heating has an influence on the bonding between the ribbons. Afterwards, investigations will be done to see what structural transformations appear in the weld area.

R E F E R E N C E S

- [1] *M. Maedaa, Y. Takahashi, M. Fukuhara, X. Wan, A. Inoue*, Materials Science and Engineering Volume 148, February 2008, pp. 141–144
- [2] *J. Kim, Y. Kawamura*, Scripta Materialia Volume 56, Issue 8, April 2007, pp.709-712
- [3] *Y. Kawamura, Y. Ohno, A. Chiba*, Material Science Forum Volume 386-388, January 2002, pp. 553-558
- [4] *Y. Kawamura, Y. Ohno*, Scripta Materialia Volume 45, July 2001, 127-132
- [5] *Y. Kawamura, Y. Ohno*, Scripta Materialia Volume 45, August 2001, pp. 279-285
- [6] *J. Kim, D. Lee, S. Shin, C. Lee*, Material Science and Engineering Volume 434, October 2006, pp. 194-201
- [7] *B. Li, Z.Y. Li, J.G. Xiong, L. Xing, D. Wang, Y. Li*, Journal of Alloys and Compounds Volume 413, March 2006, pp. 118-121
- [8] *Y. Kawahito, Y. Niwa, T. Terajima, S. Katayama*, Materials Transactions Volume 51, August 2010, pp. 1433-1463
- [9] *Y. Kawahito, T. Terajima, H. Kimura, T. Kuroda, K. Nakata, S. Katayama, A. Inoue*, Materials Science and Engineering B Volume B148, 2008, pp. 105-108
- [10] *M. Maeda, Y. Takahashi, M. Fukuhara, X. Wang, A. Inoue*, Materials Science and Engineering B Volume B148, 2008, pp. 141-144
- [11] *T. Terajima*, Journal of Alloys and Compounds Volume 536, September 2012, pp. 113-116
- [12] *T. Terajima, H. Kimura, A. Inoue*, Transactions of Joining and welding research institute, Volume 3, December 2010, pp. 310-311
- [13] *M. Nicolaescu, C. Codrean, E. Binchiciu, R. Bogdan*, Advanced Materials Research Volume 1157, February 2020, pp. 123-129
- [14] *M.M. Trexler, N.N. Thadhani*, Progress in Materials Science Volume 55, November 2010, pp. 759-839
- [15] *G. Kumar, T. Ohkubo, T. Mukai, K. Hono*, Scripta Materialia Volume 57, July 2007, pp. 173-176
- [16] *A. Inoue, W. Zhang*, Material Transactions Volume 45, 2004, pp. 584-587
- [17] *S. Pauly, G. Liu, G. Wang, U. Kuhn, N. Mattern, J. Eckert*, Acta Materialia Volume 57, October 2009, pp. 5445-5453